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PREDICTION OF MUTUAL DIFFUSION COEFFICIENTS FROM THERMAL CONDUCTIVITY DATA

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Prediction of Mutual Diffusion Coefficients
for Thermal Conductivity Data

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ABSTRACT

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The theoretical expression for thermal conductivity of a binary mixture of gases has been used to determine the mutual diffusion coefficient of the two gases. The theoretical thermal conductivity formula has also been used to obtain the temperature derivative of the diffusion coefficient at room temperature and these derivatives in turn are used to predict the diffusion coefficient at several temperatures above and below room temperature. The agreement between predicted and observed diffusion coefficients is good. The temperature derivatives have also been used to calculate thermal diffusion factors and the agreement is satisfactory considering the great sensitivity of the thermal diffusion factors to small errors in the derivatives.

Author

1. INTRODUCTION

The expression for the transport properties of gases obtained from the Chapman-Enskog theory [1] can be put into such a form that they can be considered as relations between various experimental properties. The particular relation to be studied here is the expression for thermal conductivity of a binary mixture. This expression can be used as a relation between the experimental thermal conductivity at a single temperature and the temperature derivative of the mutual diffusion coefficient of the two gases. Relations of this type are virtually independent of any force law between the molecules and therefore can be used to either calculate one property from another or as a test of the mutual consistency between the measurements of two different properties.

Holleran and Hulbert [2] derived a recursion for the collision integrals and showed how this formula can be used to convert the theoretical formulas into expressions between measurable quantities. The technique of using theoretical expressions to obtain relations between experimental quantities has been used recently to discuss the properties of several specific systems; thermal conductivity [3], viscosity [4], and diffusion and thermal diffusion [5,6]. The theoretical formula for the viscosity of a binary mixture has been used to determine mutual diffusion coefficients for both rare gases [7] and polyatomic and polar gases [8] quite successfully. The form of the formula for the viscosity of the binary mixture is quite similar to that of the thermal conductivity and thus the same procedure should be applicable to both. Therefore, experimental thermal conductivities, measured at room temperature, have been used to estimate diffusion coefficients over a range of temperatures. These derived values are compared to directly measured values thereby testing the mutual consistency of the two properties. Values of the diffusion coefficients obtained from viscosity data [7] are also compared to those obtained from thermal conductivity measurements. Furthermore, since the thermal diffusion factor is related to the temperature dependence of the diffusion coefficient [2,5] comparisons to the measured thermal diffusion factors are also made. The last comparison is an extremely severe test of the derived temperature derivatives because the thermal diffusion factor is very sensitive to small errors in the derivatives.

The specific formula for the thermal conductivity of a mixture used here has been given by Muckenfuss and Curtiss [9]. This formula is based on the Chapman-Enskog theory [10]. This theory involves several assumptions among which are the assumptions of spherically symmetric force fields and elastic molecular collisions and no allowance is made for the transport of energy by the internal degrees of freedom in the molecule. Therefore, these calculations have been limited to systems composed of noble gases.

2. Procedure

The expression for the first approximation to thermal conductivity of a binary mixture [9] can be rearranged into the form

$$(pD_{12})^2 a + (pD_{12})b + (pD_{12})B_{12}^*c + B_{12}^*d + e = 0, \quad (1)$$

where D_{12} is the mutual diffusion coefficient, p is the pressure in atmosphere and B_{12}^* is the ratio of collision integrals [1]

$$B^* = (5\Omega^{(1,2)*} - 4\Omega^{(1,3)*})/\Omega^{(1,1)*},$$

which depends on the temperature and the force law and is usually near unity, and

$$a = 25(M_1+M_2)^2 x_1 x_2 (\lambda_1 + \lambda_2 - \lambda_{mix}) / \lambda_1 \lambda_2 T, \quad (2)$$

$$b = 30(M_1 x_2 + M_2 x_1)^2 + 25(M_1 x_1 + M_2 x_2)^2 + 16 M_1 M_2 A_{12}^* (x_1 - x_2)^2 - \frac{\lambda_{mix}}{\lambda_1 \lambda_2} [30(M_1 x_2^2 \lambda_1 + M_2 x_1^2 \lambda_2) + 25(M_1 x_1^2 \lambda_2 + M_2 x_2^2 \lambda_1) + 16 M_1 M_2 A_{12}^* (x_1^2 \lambda_2 + x_2^2 \lambda_1)], \quad (3)$$

$$c = 12 \left[\frac{\lambda_{mix}}{\lambda_1 \lambda_2} (M_1 x_1^2 \lambda_2 + M_2 x_2^2 \lambda_1) - (M_1 x_1 + M_2 x_2)^2 \right], \quad (4)$$

$$d = \frac{72 x_1 x_2 T}{5(M_1 + M_2)^2} \left[(M_1^2 - M_2^2)^2 + \frac{8}{15} M_1 M_2 A_{12}^* (M_1 + M_2)^2 \right] \lambda_{mix}, \quad (5)$$

$$e = - \frac{16 x_1 x_2 T}{(M_1 + M_2)^2} \left[\frac{15}{8} (M_1^2 - M_2^2)^2 + \frac{11}{5} M_1 M_2 A_{12}^* (M_1 + M_2)^2 \right] \lambda_{mix}. \quad (6)$$

In these equations x_1 and x_2 are the mole fractions of the components, M_1 and M_2 their molecular weights, λ_1 and λ_2 the thermal conductivity of the pure components, λ_{mix} is the

thermal conductivity of the mixture, T is in $^{\circ}\text{K}$, and $R = 82.0567 \text{ cm}^3\text{-atm/mole} - ^{\circ}\text{K}$ is the gas constant. The quantity A_{12}^* is a ratio of collision integrals, $\Omega^{(2,2)}*/\Omega^{(1,1)}*$, which depends only weakly on the temperature and force law between the unlike molecules, and is usually nearly equal to unity.

Equations (1) - (6) constitute a relation between the three quantities D_{12}^* , A_{12}^* and B_{12}^* . If the thermal conductivity of a mixture at a given temperature is known for three different compositions then it should be possible in principle to obtain values for all three quantities. However, when curves of B_{12}^* vs. D_{12} for a given value of A_{12}^* are plotted for the different compositions they have about the same slope and lie close together, but they seldom intersect. Although a simultaneous solution is therefore impractical it is possible to choose values, based on experiment or realistic force laws, for two of the three quantities and determine the third from the above equations. The quantity which appears to vary least with temperature or force law is A_{12}^* . This can be seen by examination of the tables given by Hirschfelder, Curtis and Bird [1] for the 12-6 potential and the exp-6 potential. Therefore values of A_{12}^* based on the exp-6 potential and the combination rules suggested by Mason and Rice [1] were used in eqs. (2)-(5).

When a theoretical value is assigned to A_{12}^* , Eq (1) becomes a relation between D_{12} and B_{12}^* . If either quantity is known then the other can be computed. That is, a theoretical value of B_{12}^* can be used to obtain D_{12} , or an experimental value of D_{12} can be used to yield a value for B_{12}^* . The former method is very similar to the way the viscosity of a binary mixture, η_{mix} , has been used to determine D_{12} [7,8].

Both techniques have been used. However it is of particular interest to determine B_{12}^* from known values of D_{12} because it has been shown [5] that B_{12}^* is related to temperature derivative of D_{12} ,

$$B_{12}^* = \frac{1}{T^2} \left[1 - 2 \left(\frac{\partial \ln D_{12}}{\partial \ln T} \right)_P \right] \left[2 \left(\frac{\partial \ln D_{12}}{\partial \ln T} \right)_P - 9 \right] + \frac{1}{3} \left(\frac{\partial^2 \ln D_{12}}{\partial (\ln T)^2} \right)_P, \quad (7)$$

where the last term on the right side of Eq(7) can be dropped as negligible in the present case. Thus if λ_{mix} and D_{12} are known at a given temperature Eq(7) provides a means of predicting values D_{12} in the neighborhood of the given temperature. If D_{12} is known as a function of temperature then Eq(1) and (7) provide a good test of the consistency of the experimental λ_{mix} and D_{12} .

The thermal conductivity measurements of Thornton and Baker [12,13,14], von Uebisch [15], Saxena and Srivastava [16,17], have been compared to Eqs(1) and (7) using both directly measured values of D_{12} and values determined from viscosity data [7]. The λ_{mix} measured near room temperature were used along interpolated values of D_{12} to evaluate $(\partial \ln D_{12} / \partial \ln T)_p$. These derivatives and the corresponding values of D_{12} were used to determine values of D_{12} at each temperature where there were experimentally measured D_{12} and at the temperatures of the D_{12} obtained from viscosity.

3. Results

The diffusion coefficients obtained by using a theoretical value of B_{12} in Eq(1) are given in Table 1. The temperature can be used to identify the source of the λ_{mix} data. Thornton and Baker worked at $18^\circ C$, von Uebisch at $29^\circ C$ and Saxena and Srivastava at $38^\circ C$. The D_{12} given in the third column have been obtained from λ_{mix} . These are the averages of the different values obtained for each composition. The spread of the values is usually a few per cent. The accuracy of this procedure seems to decrease rapidly as the mole fraction of either component becomes very small, therefore, the results of certain dilute mixtures were neglected when they were not in good agreement with values obtained at other compositions. This disagreement is probably due to a magnification of experimental errors occurring as λ_{mix} approaches either λ_1 or λ_2 and the coefficients given by Eqs(2) to (6) become dominated by small differences between comparable quantities. One system which is particularly troublesome in this respect is Kr-Ar. The thermal conductivity data at only one composition, near $x_1=0.5$, at $302^\circ K$ and $311^\circ K$ were used. The next column gives values of D_{12} interpolated from the D_{12} obtained from viscosity data [7]. The fifth column gives

interpolated experimental D_{12} . The sources of the experimental D_{12} have been given previously [7]. The sixth column contains theoretical D_{12} which were calculated using the formula for the first approximation to the diffusion coefficient [1] and the exp-6 potential with the force constants given by Mason, Rice and von Ubisch [11,18]. The last two columns give the values of B_{12} and A_{12}^* used, these are based on the same parameters as the theoretical D_{12} .

The results given in Table 1 show that for rare gas mixtures thermal conductivity data can be used to determine diffusion coefficients as successfully as viscosity data. Since B_{12}^* depends only slightly more than A_{12}^* on the choice of force law, the use of theoretical values for both B_{12}^* and A_{12}^* in eqs (1)-(6) does not introduce the force law in a pronounced manner into the calculations. This fact was used by Saxena and Agrawal [19] to show that Thornton's viscosity and thermal conductivity measurements are mutually consistent.

A more stringent test of the congruity of the D_{12} and λ_{mix} data is to calculate the slope of the $\ln D_{12}$ vs. $\ln T$ curve at the temperature of λ_{mix} data, using Eqs (1) and (7), and then extrapolating to the temperatures where D_{12} is known. The results of these calculations are given in Table 2. The table is in two parts. The first part gives results based on Eq (1) and (7) and the experimental D_{12} listed previously in Table 1. The second part gives the results using the D_{12} based on viscosity data; these D_{12} are also listed in Table 1. The third column contains the observed D_{12} given in order of increasing temperature, first are the directly measured values followed by the values determined from γ_{mix} . The sources of the experimental D_{12} have been given before [7]. The next columns list the results based on λ_{mix} data followed in each case by the percentage deviation. The source of the λ_{mix} data can be identified by the column headings which give the temperature of the λ_{mix} measurements: 18°C , Thornton and Baker [12,13,14], 29°C , von Ubisch [15], and 38° , Saxena and Srivastava [16,17]. Thornton and Baker have given two sets of measurements for Ar-He, but only the results for the set (Table 2, ref. 12) which gives the best agreement with other values of D_{12} has been included in Table 2.

The results in Table 1 and 2 can be used to discuss the mutual consistency of the D_{12} determined from λ mix and the D_{12} obtained from other sources. The agreement between the measured thermal conductivities by the various workers and the agreement with theoretical calculations has been discussed by Thornton and Baker [13,14], Saxena and Gandhi [20] and Mason and von Ubsich [18].

For the Ne-He system the experimental data of Thornton and Baker and von Ubsich are not in satisfactory agreement [14] with each other. The D_{12} based on von Ubsich's data are in better agreement with experimental D_{12} and especially with the values derived from η mix. The indication is that there is a slight incongruity between η mix and λ mix on the one hand and the experiment D_{12} on the other hand.

The measurements of the Ar-He system exhibit a high degree of internal consistency, when Thornton and Baker's first set of λ mix data are ignored.

The temperature range of the measured D_{12} for Kr-He is limited and therefore mutual consistency appears to be very good.

The results for the Xe-He systems reflect the discrepancy between the experimental D_{12} and the D_{12} derived from mix. There is uniform disagreement between the theoretical values of λ mix and the experimental values [12,18,20] as well as between the theoretical and experimental values of mix [12]. This discordance has been attributed to the intermolecular potential chosen for Xe and Xe-He [20]. The experimental D_{12} [21] are in good agreement with theoretical calculations. Since the derived values of D_{12} which are based on two different kinds of data measured by several different investigators, are in good agreement with each other, it seems as if the experimental D_{12} should be reinvestigated.

The results for Ar-Ne show that the slope obtained from von Ubsich's data agrees best with the experimental D_{12} and the combination of Saxena's data and a value of D_{12} based on viscosity data yields the best over all agreement. The general consistency between λ mix and the D_{12} is only fair.

The same remarks made for the Kr-He system apply to the Kr-Ne and the Xe-Ne systems.

The results for Kr-Ar are similar to those of Ar-Ne. Thermal conductivities measured by von Übisch show the highest consistency with the experimental D_{12} , but the calculations are based on λ mix at only one composition.

For the Xe-Kr system the consistency seems to be good for all three λ mix measurements, with those of von Übisch appearing best.

There is not enough independently determined D_{12} to test the Xe-Kr system.

On the basis of the results given in Table 2 it appears as if the slopes of the $\ln D_{12}$ vs $\ln T$ curves derived from λ mix could be used to predict values of D_{12} for those systems for which D_{12} is known at a few temperatures. The predicted values would be expected to be accurate to better than 10% over range of about 100° above and below room temperature. There is no reason to recommend the slope based on a particular set of λ mix over the others for these systems, therefore all the calculated slopes are listed in Table 3 for those systems for which there are limited data for D_{12} .

There is another extremely sensitive test for the calculated slopes. This is to use the slopes to calculate the magnitude of the thermal diffusion factor. The thermal diffusion factor, α_T , can be written in terms of experimental quantities as [5],

$$\alpha_T = 2[2 - (\partial \ln D / \partial \ln T)p] (-S_2/Q_2) , \quad (8)$$

where, in terms of the thermal conductivity,

$$-S_2 = \frac{15M_1(M_1-M_2)}{2(M_1+M_2)^2} + \frac{4M_1M_2A_{12}^*}{(M_1+M_2)^2} - \frac{25}{4} \frac{M_2}{M_1+M_2} \frac{pD_{12}}{T\lambda_2} \quad (9)$$

$$Q_2 = \frac{25}{2} \cdot \frac{1}{(M_1 + M_2)^2} \cdot \frac{p D_{12}}{T \lambda_2} \cdot \left[3M_1^2 + M_2^2 + \frac{8}{5} M_1 M_2 A_{12}^* \right]. \quad (10)$$

The α_T calculated from Eq.(8) are very sensitive to small errors in the slope due to the fact that the slope is usually not far from 2. Eq.(8) is the expression for the first approximation to the thermal diffusion factor for the case when the heavy gas is present as a trace. There are several reasons for considering this particular case. The expression for the first approximation is simpler than at intermediate compositions and the errors due to neglecting higher approximations are smallest for this composition [22]. Furthermore there are some experimental measurements at this composition for Ar-He [23] and Xe-He, Xe-Ne, and Xe-Ar [24].

In Table 4 are given the experimental values of α_T , calculated from the smoothed expressions [23, 24]. Also given are values of α_T calculated with Eq.(8) using the experimental D_{12} in Eqs.(9) and (10) and the $(\partial \ln D_{12} / \partial \ln T)p$ obtained from Eqs.(1) and (7) based on the experimental D_{12} , and the values of α_T obtained in the same manner using the D_{12} obtained from η_{mix} data. The agreement for Ar-Ne and Xe-Ne is surprisingly good considering the sensitivity of α_T to $(\partial \ln D_{12} / \partial \ln T)p$. The results for Xe-He indicate that the D_{12} based on viscosities are more consistent with the experimental α_T than the measured D_{12} . This indication is in agreement with the conclusion reached earlier.

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Table I

Diffusion Coefficients Obtained from λ_{mix} using
Theoretical Values of B_{12}^*

| System | T°K | pD ₁₂ atm - cm ² /sec | | | | | |
|--------|-----|---|--------------------------|--------|--------|------------|------------|
| | | from λ_{mix} | from η_{mix} | exptl. | theor. | B_{12}^* | A_{12}^* |
| Ne-He | 291 | 1.21 | 1.05 | 1.00 | 1.01 | 1.115 | 1.144 |
| | 302 | 1.12 | 1.12 | 1.06 | 1.08 | 1.121 | 1.145 |
| Ar-He | 291 | 0.778 | 0.708 | 0.718 | 0.706 | 1.116 | 1.131 |
| | 291 | 0.713 | 0.708 | 0.718 | 0.706 | 1.116 | 1.131 |
| | 302 | 0.772 | 0.754 | 0.765 | 0.753 | 1.117 | 1.132 |
| | 311 | 0.782 | 0.792 | 0.805 | 0.791 | 1.117 | 1.133 |
| Kr-He | 291 | 0.650 | 0.616 | 0.618 | 0.633 | 1.115 | 1.126 |
| | 302 | 0.662 | (0.659) | 0.658 | 0.675 | 1.115 | 1.127 |
| Xe-He | 291 | 0.531 | 0.513 | 0.561 | 0.553 | 1.118 | 1.126 |
| | 302 | 0.548 | 0.546 | 0.599 | 0.619 | 1.118 | 1.127 |
| | 311 | 0.588 | 0.573 | 0.631 | 0.652 | 1.118 | 1.123 |
| Ar-Ne | 291 | 0.326 | 0.301 | 0.310 | 0.308 | 1.106 | 1.104 |
| | 302 | 0.339 | 0.321 | 0.332 | 0.328 | 1.105 | 1.105 |
| | 311 | 0.342 | 0.338 | 0.350 | 0.345 | 1.105 | 1.106 |
| Kr-Ne | 291 | 0.249 | 0.252 | 0.247 | 0.243 | 1.118 | 1.100 |
| | 302 | 0.273 | (0.268) | 0.262 | 0.260 | 1.118 | 1.100 |
| | 311 | 0.276 | (0.281) | 0.275 | 0.274 | 1.118 | 1.100 |
| Xe-Ne | 291 | 0.212 | 0.208 | 0.207 | 0.220 | 1.122 | 1.102 |
| | 302 | 0.231 | (0.222) | 0.221 | 0.236 | 1.120 | 1.102 |
| Kr-Ar | 291 | 0.126 | 0.132 | 0.136 | 0.132 | 1.135 | 1.101 |
| | 302 | 0.148 | (0.143) | 0.145 | 0.141 | 1.133 | 1.101 |
| | 311 | 0.150 | (0.152) | 0.153 | 0.149 | 1.131 | 1.101 |
| Xe-Ar | 291 | 0.105 | 0.107 | 0.107 | 0.110 | 1.145 | 1.103 |
| | 302 | 0.118 | (0.114) | 0.115 | 0.118 | 1.142 | 1.103 |
| | 311 | 0.119 | (0.119) | 0.122 | 0.125 | 1.140 | 1.103 |
| Xe-Kr | 291 | 0.0721 | 0.0725 | | 0.0728 | 1.170 | 1.108 |
| | 302 | 0.0762 | (0.0773) | | 0.0782 | 1.166 | 1.107 |

The values in parenthesis are extrapolated values using the slope and value determined at 291°K

Table 2a

Comparison of diffusion coefficients obtained from λ_{mix} and one experimental D_{12} with measured D_{12} and D_{12} obtained from η_{mix}

| | | $pD_{12} \text{ atm- cm}^2/\text{sec}$ | | | | | | |
|--------|-------|--|--------|-------|--------|-------|-------|------|
| System | T°K | obs. | 18°C | dev | 29°C | dev | 38°C | dev |
| Ne-He | 273 | 0.906 | 0.822 | -9.3 | 0.876 | -3.4 | | |
| | 288 | 0.986 | 0.970 | -1.6 | 0.971 | -1.6 | | |
| | 303 | 1.065 | 1.13 | 6.6 | 1.07 | 0.5 | | |
| | 318 | 1.158 | 1.32 | 13.8 | 1.17 | 1.4 | | |
| | 20.4 | 0.0102 | 0.0003 | -97.4 | 0.0059 | -42.0 | | |
| | 65.8 | 0.0860 | 0.0101 | -88.3 | 0.0564 | -34.4 | | |
| | 90.1 | 0.144 | 0.027 | -81.5 | 0.103 | -28.2 | | |
| | 90.2 | 0.146 | 0.027 | -81.7 | 0.104 | -29.0 | | |
| | 194.0 | 0.532 | 0.286 | -46.3 | 0.453 | -14.8 | | |
| | 291.2 | 1.06 | 1.00 | -5.3 | 0.991 | -6.5 | | |
| | 293.2 | 1.05 | 1.03 | -2.4 | 1.00 | -4.3 | | |
| | 293.2 | 1.10 | 1.03 | -6.8 | 1.00 | -8.7 | | |
| | 293.2 | 1.06 | 1.03 | -3.3 | 1.00 | -5.2 | | |
| | 373.2 | 1.60 | 2.16 | 35.2 | 1.59 | -0.1 | | |
| | 473.2 | 2.37 | 4.51 | 90.3 | 2.53 | 6.6 | | |
| | 523.2 | 2.78 | 6.15 | 121.4 | 3.07 | 10.3 | | |
| Ar-He | 251 | 0.544 | 0.561 | 3.1 | 0.557 | 2.4 | 0.568 | 4.4 |
| | 259 | 0.583 | 0.591 | 1.3 | 0.588 | 0.8 | 0.598 | 2.5 |
| | 269 | 0.639 | 0.629 | -1.5 | 0.627 | -1.9 | 0.635 | -0.6 |
| | 273 | 0.640 | 0.645 | 0.8 | 0.643 | 0.5 | 0.651 | 1.7 |
| | 280 | 0.703 | 0.673 | -4.3 | 0.672 | -4.5 | 0.678 | -3.5 |
| | 288 | 0.701 | 0.705 | 0.6 | 0.705 | 0.6 | 0.710 | 1.3 |
| | 289 | 0.731 | 0.709 | -3.0 | 0.709 | -3.0 | 0.714 | -2.3 |
| | 296 | 0.744 | 0.738 | -0.8 | 0.739 | -0.7 | 0.742 | -0.2 |
| | 303 | 0.760 | 0.767 | 1.0 | 0.769 | 1.2 | 0.771 | 1.4 |
| | 318 | 0.825 | 0.832 | 0.8 | 0.835 | 1.3 | 0.834 | 1.1 |
| | 328 | 0.851 | 0.876 | 2.9 | 0.881 | 3.5 | 0.877 | 3.0 |
| | 337 | 0.889 | 0.916 | 3.1 | 0.923 | 3.8 | 0.916 | 3.1 |
| | 354 | 1.000 | 0.995 | -0.5 | 1.00 | 0.0 | 0.992 | -0.7 |

| | | pD ₁₂ atm - cm ² /sec | | | | | | |
|--------|-------|---|--------|-------|--------|------|--------|------|
| System | T°K | obs | 18°C | dev | 29°C | dev | 38°C | dev |
| Ar-He | 366 | 1.060 | 1.05 | -0.8 | 1.06 | 0.0 | 1.05 | -1.1 |
| | 372 | 1.122 | 1.08 | -3.7 | 1.09 | -2.6 | 1.08 | -4.1 |
| | 387 | 1.165 | 1.15 | -0.9 | 1.17 | 0.4 | 1.15 | -1.5 |
| | 405 | 1.288 | 1.24 | -3.3 | 1.26 | -1.8 | 1.24 | -4.1 |
| | 418 | 1.344 | 1.31 | -2.6 | 1.33 | -0.7 | 1.30 | -3.3 |
| | 72.0 | 0.0681 | 0.0691 | 1.4 | 0.0653 | -4.2 | 0.0745 | 9.5 |
| | 81.0 | 0.0823 | 0.0841 | 2.2 | 0.0799 | -2.9 | 0.0903 | 9.7 |
| | 90.2 | 0.102 | 0.101 | -1.0 | 0.096 | -5.8 | 0.107 | 5.4 |
| | 192.5 | 0.349 | 0.359 | 2.8 | 0.353 | 1.1 | 0.369 | 5.7 |
| | 229.5 | 0.484 | 0.482 | -0.4 | 0.477 | -1.4 | 0.491 | 1.4 |
| | 291.1 | 0.730 | 0.718 | -1.6 | 0.718 | -1.6 | 0.722 | -1.1 |
| | 291.2 | 0.698 | 0.718 | 2.9 | 0.718 | 2.9 | 0.723 | 3.5 |
| | 293.2 | 0.708 | 0.726 | 2.6 | 0.727 | 2.7 | 0.731 | 3.2 |
| | 373.2 | 1.07 | 1.09 | 1.7 | 1.10 | 2.8 | 1.08 | 1.1 |
| | 473.2 | 1.59 | 1.62 | 1.9 | 1.65 | 4.0 | 1.59 | 0.0 |
| Kr-He | 273 | 0.556 | 0.551 | -1.0 | 0.554 | -0.4 | | |
| | 288 | 0.605 | 0.606 | 0.2 | 0.606 | 0.2 | | |
| | 303 | 0.659 | 0.664 | 0.8 | 0.661 | 0.3 | | |
| | 318 | 0.720 | 0.724 | 0.6 | 0.718 | -0.3 | | |
| | 291.2 | 0.616 | 0.618 | 0.3 | 0.618 | 0.3 | | |
| Xe-He | 273 | 0.501 | 0.507 | 1.1 | 0.513 | 2.5 | 0.515 | 2.7 |
| | 288 | 0.550 | 0.552 | 0.3 | 0.557 | 1.3 | 0.559 | 1.7 |
| | 303 | 0.604 | 0.598 | -1.0 | 0.602 | -0.3 | 0.606 | 0.3 |
| | 318 | 0.655 | 0.646 | -1.4 | 0.648 | -1.1 | 0.653 | -0.3 |
| | 291.3 | 0.512 | 0.562 | 9.7 | 0.567 | 10.7 | 0.569 | 11.2 |
| | 293. | 0.518 | 0.567 | 9.5 | 0.572 | 10.4 | 0.575 | 10.9 |
| | 400 | 0.880 | 0.931 | 5.7 | 0.920 | 4.5 | 0.935 | 6.2 |
| | 500 | 1.27 | 1.33 | 4.4 | 1.29 | 1.9 | 1.33 | 4.4 |
| | 550 | 1.49 | 1.54 | 3.5 | 1.50 | 0.4 | 1.54 | 3.5 |
| Ar-Ne | 90 | 0.0360 | 0.0189 | -47.4 | 0.0343 | -4.9 | 0.0567 | 57.5 |
| | 194.5 | 0.159 | 0.119 | -25.3 | 0.145 | -8.6 | 0.176 | 10.5 |
| | 273 | 0.285 | 0.266 | -6.6 | 0.274 | -3.7 | 0.289 | 1.4 |

pD₁₂ atm - cm²/sec

| System | T°K | obs | 18°C | dev | 29°C | dev | 38°C | dev |
|--------|-------|--------|--------|-------|--------|-------|--------|-------|
| Ar-Ne | 273 | 0.276 | 0.266 | -3.5 | 0.274 | -0.6 | 0.289 | 4.7 |
| | 288 | 0.300 | 0.302 | 0.8 | 0.303 | 1.0 | 0.313 | 4.2 |
| | 303 | 0.327 | 0.341 | 4.4 | 0.334 | 2.1 | 0.337 | 3.0 |
| | 318 | 0.357 | 0.383 | 7.3 | 0.365 | 2.3 | 0.362 | 1.2 |
| | 353 | 0.442 | 0.491 | 11.1 | 0.444 | 0.5 | 0.421 | -4.7 |
| | 473 | 0.738 | 0.986 | 33.6 | 0.769 | 4.2 | 0.647 | -12.3 |
| | 72.3 | 0.0259 | 0.0104 | -60.0 | 0.0216 | -16.5 | 0.0408 | 57.7 |
| | 90.3 | 0.0386 | 0.0178 | -53.8 | 0.0331 | -14.3 | 0.0566 | 46.7 |
| | 193.4 | 0.147 | 0.114 | -22.2 | 0.142 | -3.7 | 0.174 | 18.3 |
| | 229.0 | 0.205 | 0.173 | -15.7 | 0.196 | -4.6 | 0.223 | 8.7 |
| | 291.1 | 0.311 | 0.310 | -0.2 | 0.309 | -0.6 | 0.317 | 2.1 |
| | 291.2 | 0.292 | 0.311 | 6.4 | 0.309 | 6.0 | 0.318 | 8.7 |
| | 293.2 | 0.309 | 0.316 | 2.2 | 0.314 | 1.4 | 0.321 | 3.8 |
| | 373.2 | 0.466 | 0.569 | 22.1 | 0.497 | 6.6 | 0.458 | -1.8 |
| | 473.2 | 0.695 | 1.02 | 46.1 | 0.782 | 12.5 | 0.649 | -6.6 |
| | 523.2 | 0.822 | 1.30 | 57.8 | 0.947 | 15.2 | 0.752 | -8.5 |
| Kr-Ne | 273 | 0.223 | 0.221 | -0.8 | 0.217 | -2.6 | 0.220 | -1.3 |
| | 288 | 0.240 | 0.242 | 0.9 | 0.240 | 0.0 | 0.241 | 0.4 |
| | 303 | 0.266 | 0.264 | -0.7 | 0.263 | -1.0 | 0.263 | -1.0 |
| | 318 | 0.284 | 0.287 | 1.0 | 0.288 | 1.5 | 0.285 | 0.5 |
| | 291.2 | 0.252 | 0.247 | -2.0 | 0.245 | -2.9 | 0.246 | -2.6 |
| Xe-Ne | 273 | 0.186 | 0.185 | -0.5 | 0.184 | -1.0 | | |
| | 288 | 0.202 | 0.203 | 0.5 | 0.203 | 0.5 | | |
| | 303 | 0.221 | 0.222 | 0.6 | 0.222 | 0.6 | | |
| | 318 | 0.244 | 0.242 | -0.6 | 0.242 | -0.6 | | |
| | 291.2 | 0.208 | 0.207 | -0.5 | 0.207 | -0.5 | | |
| Kr-Ar | 199.5 | 0.0720 | 0.0896 | 23.6 | 0.0653 | -9.3 | 0.0773 | 7.3 |
| | 273 | 0.126 | 0.127 | 0.8 | 0.120 | -5.2 | 0.125 | -0.7 |
| | 273 | 0.119 | 0.127 | 6.7 | 0.120 | 0.4 | 0.125 | 5.2 |
| | 288 | 0.128 | 0.134 | 4.7 | 0.132 | 3.5 | 0.136 | 6.1 |
| | 303 | 0.140 | 0.142 | 1.5 | 0.146 | 4.3 | 0.147 | 4.9 |

| | | pD ₁₂ atm- cm ² /sec | | | | | | |
|--------|-------|--|--------|-------|--------|------|--------|-------|
| System | T°K | obs | 18°C | dev | 29°C | dev | 38°C | dev |
| Kr-Ar | 318 | 0.153 | 0.150 | -1.9 | 0.160 | 4.7 | 0.158 | 3.4 |
| | 353 | 0.197 | 0.168 | -14.6 | 0.196 | -0.5 | 0.186 | -5.7 |
| | 373 | 0.216 | 0.179 | -17.1 | 0.218 | 0.8 | 0.202 | -6.4 |
| | 473 | 0.327 | 0.232 | -29.0 | 0.344 | 5.3 | 0.291 | -10.9 |
| | 291.2 | 0.132 | 0.136 | 3.0 | 0.135 | 2.4 | 0.138 | 4.6 |
| Xe-Ar | 194.7 | 0.0508 | 0.0555 | 9.3 | 0.0507 | -0.1 | 0.0562 | 10.5 |
| | 273 | 0.0943 | 0.0966 | 2.4 | 0.0952 | 1.0 | 0.0980 | 3.9 |
| | 273.2 | 0.0962 | 0.0967 | 0.5 | 0.0953 | -0.9 | 0.0981 | 2.0 |
| | 288 | 0.102 | 0.105 | 3.2 | 0.105 | 3.2 | 0.107 | 4.9 |
| | 303 | 0.114 | 0.114 | 0.0 | 0.116 | 1.4 | 0.116 | 2.1 |
| | 318 | 0.128 | 0.124 | -3.2 | 0.126 | -1.2 | 0.126 | -1.6 |
| | 329.9 | 0.1366 | 0.132 | -3.6 | 0.136 | -0.8 | 0.134 | -2.0 |
| | 329.9 | 0.1373 | 0.132 | -4.1 | 0.136 | -1.3 | 0.134 | -2.5 |
| | 378.0 | 0.1759 | 0.165 | -6.4 | 0.175 | -0.7 | 0.168 | -4.7 |
| | 291.2 | 0.107 | 0.107 | 0.0 | 0.107 | 0.0 | 0.109 | 1.8 |

Table 2b

Comparison of diffusion coefficients obtained from λ_{mix} and one D_{12} determined from viscosity data with experimental D_{12} and D_{12} obtained from η_{mix} .

| System | T°K | obs | $pD_{12} \text{ atm} \cdot \text{cm}^2/\text{sec}$ | | | | |
|--------|-------|--------|--|-------|--------|-------|-------|
| | | | 18°C | dev | 29°C | dev | 38°C |
| Ne-He | 273 | 0.906 | 0.891 | -1.7 | 0.944 | 4.2 | |
| | 288 | 0.986 | 1.02 | 3.9 | 1.03 | 4.9 | |
| | 303 | 1.065 | 1.17 | 9.8 | 1.13 | 6.0 | |
| | 318 | 1.158 | 1.33 | 14.5 | 1.23 | 5.8 | |
| | 20.4 | 0.0102 | 0.0010 | -89.9 | 0.0112 | -10.1 | |
| | 65.8 | 0.0860 | 0.0218 | -74.6 | 0.0831 | -3.4 | |
| | 90.1 | 0.144 | 0.050 | -66.0 | 0.142 | -1.3 | |
| | 90.2 | 0.146 | 0.050 | -66.0 | 0.142 | -2.5 | |
| | 194.0 | 0.532 | 0.366 | -31.3 | 0.527 | -1.0 | |
| | 291.2 | 1.06 | 1.05 | -0.5 | 1.05 | -0.5 | |
| | 293.2 | 1.05 | 1.07 | 2.2 | 1.07 | 1.6 | |
| | 293.2 | 1.10 | 1.07 | -2.2 | 1.07 | 3.0 | |
| | 293.2 | 1.06 | 1.07 | 1.2 | 1.07 | 0.6 | |
| Ar-He | 373.2 | 1.60 | 2.01 | 25.8 | 1.61 | 0.7 | |
| | 473.2 | 2.37 | 3.74 | 57.7 | 2.42 | 2.0 | |
| | 523.2 | 2.78 | 4.86 | 74.7 | 2.87 | 3.2 | |
| | 251 | 0.544 | 0.550 | 1.0 | 0.544 | 0.0 | 0.544 |
| | 259 | 0.583 | 0.580 | -0.5 | 0.575 | -1.4 | 0.583 |
| | 269 | 0.639 | 0.619 | -3.2 | 0.615 | -3.8 | 0.621 |
| | 273 | 0.640 | 0.635 | -0.8 | 0.631 | -1.4 | 0.637 |
| | 280 | 0.703 | 0.663 | -5.7 | 0.659 | -6.2 | 0.664 |
| | 288 | 0.701 | 0.695 | -0.8 | 0.693 | -1.1 | 0.696 |
| | 289 | 0.731 | 0.700 | -4.2 | 0.697 | -4.6 | 0.700 |
| | 296 | 0.744 | 0.729 | -2.1 | 0.727 | -2.3 | 0.729 |
| | 303 | 0.760 | 0.758 | -0.2 | 0.757 | -0.3 | 0.757 |
| | 318 | 0.825 | 0.824 | -0.1 | 0.824 | -0.1 | 0.821 |
| | | | | | | | -0.5 |

Table 2b Cont'd.

pD_{12} atm - cm^2/sec

| System | T°K | obs | 18°C | dev | 29°C | dev | 38°C | dev |
|--------|-------|--------|--------|-------|--------|-------|--------|------|
| Xe-He | 500 | 1.27 | 1.33 | 5.0 | 1.29 | 1.3 | 1.31 | 3.4 |
| | 550 | 1.49 | 1.58 | 5.9 | 1.51 | 1.6 | 1.55 | 4.2 |
| Ar-Ne | 90 | 0.0360 | 0.0067 | -81.4 | 0.0146 | -59.6 | 0.0372 | 3.4 |
| | 194.5 | 0.159 | 0.081 | -48.8 | 0.104 | -34.4 | 0.147 | -7.8 |
| | 273 | 0.285 | 0.244 | -14.3 | 0.248 | -13.0 | 0.268 | -6.0 |
| | 273 | 0.276 | 0.244 | -11.5 | 0.248 | -10.2 | 0.268 | -2.9 |
| | 288 | 0.300 | 0.291 | -3.1 | 0.284 | -5.3 | 0.295 | -1.8 |
| | 303 | 0.327 | 0.343 | 4.8 | 0.324 | -1.1 | 0.322 | -1.4 |
| | 318 | 0.357 | 0.401 | 12.2 | 0.366 | 2.5 | 0.352 | -1.5 |
| | 353 | 0.442 | 0.562 | 27.1 | 0.478 | 8.1 | 0.423 | -4.3 |
| | 473 | 0.738 | 1.45 | 96.6 | 1.01 | 36.8 | 0.712 | -3.5 |
| | 72.3 | 0.0259 | 0.0033 | -87.3 | 0.0083 | -67.9 | 0.0252 | -2.6 |
| | 90.3 | 0.0386 | 0.0068 | -82.5 | 0.0147 | -62.0 | 0.0375 | -3.0 |
| | 193.4 | 0.147 | 0.080 | -45.6 | 0.103 | -30.1 | 0.145 | -1.3 |
| | 229.0 | 0.205 | 0.138 | -32.6 | 0.158 | -22.8 | 0.196 | -4.4 |
| | 291.1 | 0.311 | 0.301 | -3.3 | 0.292 | -6.1 | 0.300 | -3.4 |
| | 291.2 | 0.292 | 0.301 | 3.1 | 0.292 | 0.0 | 0.301 | 2.9 |
| | 293.2 | 0.309 | 0.308 | -0.3 | 0.297 | -3.7 | 0.304 | -1.6 |
| | 373.2 | 0.466 | 0.673 | 44.4 | 0.551 | 18.2 | 0.467 | 0.3 |
| | 473.2 | 0.695 | 1.45 | 109.0 | 1.01 | 45.4 | 0.713 | 2.5 |
| | 523.2 | 0.822 | 2.01 | 144.7 | 1.31 | 58.9 | 0.852 | 3.7 |
| Kr-Ne | 273 | 0.223 | 0.227 | 1.7 | 0.224 | 0.4 | 0.227 | 1.7 |
| | 288 | 0.240 | 0.248 | 3.2 | 0.246 | 2.5 | 0.248 | 3.2 |
| | 303 | 0.266 | 0.269 | 1.2 | 0.269 | 1.2 | 0.269 | 1.2 |
| | 318 | 0.284 | 0.292 | 2.6 | 0.293 | 3.2 | 0.291 | 2.5 |
| | 291.2 | 0.252 | 0.252 | 0.0 | 0.251 | -0.4 | 0.252 | 0.0 |
| Xe-Ne | 273 | 0.186 | 0.186 | 0.0 | 0.185 | -0.5 | | |
| | 288 | 0.202 | 0.204 | 1.0 | 0.204 | 1.0 | | |
| | 303 | 0.221 | 0.223 | 1.0 | 0.223 | 1.0 | | |
| | 318 | 0.244 | 0.243 | -0.4 | 0.244 | 0.0 | | |
| | 291.2 | 0.208 | 0.208 | 0.0 | 0.208 | 0.0 | | |

Table 2b Cont'd.

$$pD_{12} \text{ atm} \cdot \text{cm}^2/\text{sec}$$

| System | T°K | obs | 18°C | dev | 29°C | dev | 38°C | dev |
|--------|-------|--------|--------|------|--------|-------|--------|------|
| Ar-He | 328 | 0.851 | 0.868 | 2.0 | 0.870 | 2.3 | 0.864 | 1.6 |
| | 337 | 0.889 | 0.910 | 2.3 | 0.913 | 2.7 | 0.904 | 1.7 |
| | 354 | 1.000 | 0.989 | -1.1 | 0.995 | -0.5 | 0.981 | -1.9 |
| | 366 | 1.060 | 1.05 | -1.0 | 1.06 | 0.0 | 1.04 | -2.1 |
| | 372 | 1.122 | 1.08 | -4.0 | 1.09 | -3.2 | 1.07 | -5.0 |
| | 387 | 1.165 | 1.15 | -1.1 | 1.16 | -0.1 | 1.14 | -2.3 |
| | 405 | 1.288 | 1.25 | -3.3 | 1.26 | -2.2 | 1.23 | -4.7 |
| | 418 | 1.344 | 1.31 | -2.2 | 1.33 | 0.9 | 1.29 | -3.7 |
| | 72.0 | 0.0681 | 0.0650 | -4.5 | 0.0608 | -10.7 | 0.0692 | 1.7 |
| | 81.0 | 0.0823 | 0.0795 | -3.4 | 0.0748 | -9.1 | 0.0842 | 2.4 |
| | 90.2 | 0.102 | 0.096 | -6.3 | 0.090 | -11.4 | 0.101 | -1.2 |
| | 192.5 | 0.349 | 0.349 | 0.0 | 0.342 | -2.1 | 0.356 | 2.0 |
| | 229.5 | 0.484 | 0.472 | -2.5 | 0.465 | -3.9 | 0.477 | -1.5 |
| | 291.1 | 0.730 | 0.708 | -3.0 | 0.706 | -3.3 | 0.708 | -3.0 |
| | 291.2 | 0.698 | 0.709 | 1.5 | 0.706 | 1.2 | 0.709 | 1.5 |
| | 293.2 | 0.708 | 0.717 | 1.3 | 0.715 | 1.0 | 0.717 | 1.3 |
| Kr-He | 373.2 | 1.07 | 1.08 | 1.2 | 1.09 | 2.0 | 1.07 | 0.0 |
| | 473.2 | 1.59 | 1.62 | 2.2 | 1.66 | 4.2 | 1.59 | 0.0 |
| | 273 | 0.556 | 0.548 | -1.4 | 0.554 | -0.3 | | |
| | 288 | 0.605 | 0.604 | -0.2 | 0.607 | 0.3 | | |
| | 303 | 0.659 | 0.662 | 0.4 | 0.662 | 0.4 | | |
| Xe-He | 318 | 0.720 | 0.722 | 0.3 | 0.719 | -0.2 | | |
| | 291.2 | 0.616 | 0.616 | 0.0 | 0.619 | 0.4 | | |
| | 273 | 0.501 | 0.458 | -8.7 | 0.459 | -8.3 | 0.456 | -8.9 |
| | 288 | 0.550 | 0.503 | -8.6 | 0.503 | -8.6 | 0.501 | -8.9 |
| | 303 | 0.604 | 0.550 | -8.9 | 0.548 | -9.2 | 0.547 | -9.4 |
| | 318 | 0.655 | 0.600 | -8.5 | 0.595 | -9.1 | 0.596 | -9.1 |
| | 291.3 | 0.512 | 0.513 | 0.2 | 0.513 | 0.2 | 0.511 | -0.2 |
| | 293 | 0.518 | 0.519 | 0.1 | 0.518 | 0.0 | 0.516 | -0.4 |
| | 400 | 0.880 | 0.899 | 2.2 | 0.880 | 0.0 | 0.890 | 1.1 |

Table 2b Cont'd.

| System | T°K | obs | pD ₁₂ atm - cm ² /sec | | | | | |
|--------|-------|--------|---|-------|--------|------|--------|------|
| | | | 18°C | dev | 29°C | dev | 38°C | dev |
| Kr-Ar | 199.5 | 0.0720 | 0.0795 | 10.4 | 0.0565 | 21.6 | 0.0742 | 3.0 |
| | 273 | 0.126 | 0.121 | -4.0 | 0.114 | -9.7 | 0.123 | -2.4 |
| | 273 | 0.119 | 0.121 | 1.7 | 0.114 | -4.4 | 0.123 | 3.3 |
| | 288 | 0.128 | 0.130 | 1.6 | 0.128 | 0.0 | 0.134 | 4.7 |
| | 303 | 0.140 | 0.139 | -0.7 | 0.144 | 2.6 | 0.145 | 3.9 |
| | 318 | 0.153 | 0.149 | -2.6 | 0.160 | 4.5 | 0.157 | 2.7 |
| | 353 | 0.197 | 0.171 | -13.2 | 0.202 | 2.5 | 0.186 | -5.6 |
| | 373 | 0.216 | 0.184 | -14.8 | 0.228 | 5.7 | 0.203 | -5.9 |
| | 473 | 0.327 | 0.253 | -22.6 | 0.388 | 18.7 | 0.298 | -8.9 |
| | 291.5 | 0.132 | 0.132 | 0.0 | 0.131 | -0.5 | 0.136 | 3.3 |
| Xe-Ar | 194.7 | 0.0508 | 0.0551 | 8.4 | 0.0481 | -5.4 | 0.0530 | 4.3 |
| | 273 | 0.0943 | 0.0962 | 2.0 | 0.0932 | -1.1 | 0.0952 | 0.9 |
| | 273.2 | 0.0962 | 0.0963 | 0.1 | 0.0934 | -2.9 | 0.0953 | -0.9 |
| | 288 | 0.102 | 0.105 | 3.0 | 0.104 | 2.0 | 0.104 | 2.0 |
| | 303 | 0.114 | 0.114 | 0.0 | 0.114 | 0.0 | 0.114 | 0.0 |
| | 318 | 0.128 | 0.124 | -3.3 | 0.126 | -1.8 | 0.124 | -3.3 |
| | 329.9 | 0.1366 | 0.132 | -3.6 | 0.135 | -1.1 | 0.132 | -3.6 |
| | 329.9 | 0.1373 | 0.132 | -4.1 | 0.135 | -1.6 | 0.132 | -3.6 |
| | 378.0 | 0.1759 | 0.165 | -6.4 | 0.177 | 0.3 | 0.167 | -4.9 |
| | 291.2 | 0.107 | 0.107 | 0.0 | 0.106 | -1.0 | 0.106 | -1.0 |
| Xe-Kr | 291.2 | 0.0725 | 0.0725 | 0.0 | 0.0711 | -1.9 | | |

Table 3

Calculated and experimental slopes of
the $\ln D$ vs $\ln T$ curve

from λ_{mix}

| System | 18°C | 29°C | 38°C | exptl. |
|--------|----------------------|----------------------|----------------------|--------|
| Kr-He | 1.80 | 1.70 | | 1.69 |
| Kr-Ne | 1.65 | 1.77 | 1.63 | 1.63 |
| Xe-Ne | 1.76 | 1.81 | | 1.78 |
| Xe-Kr | 1.73 | 1.48 | | |

Table 4

Observed and calculated α_T

| System | $T^{\circ K}$ | α_T exptl. | α_T exptl D_{12} | η_{mix} |
|--------------------|---------------|----------------------|------------------------------|--------------|
| Ar-He | 291 | .669 | .687 | .624 |
| | 302 | .671 | .603 | .529 |
| | 311 | .672 | .797 | .726 |
| Xe-He ^a | 291 | .96 | 1.21 | .75 |
| | 302 | .97 | 1.40 | .96 |
| | 311 | .97 | 1.29 | .82 |
| Xe-Ne ^a | 291 | .57 | .51 | .54 |
| | 302 | .57 | .43 | .45 |
| Xe-Ar ^a | 291 | .19 | .45 | .43 |
| | 302 | .19 | .14 | .05 |
| | 311 | .21 | .43 | .34 |

^a These measurements refer to ^{133}Xe